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(54) Title: POLYSILOXANE COMPOSITIONS WHICH CROSS-LINK BY CONDENSATION, A PROCESS FOR THEIR PRODUCTION AND THEIR USE.

(54) Bezeichnung: KONDENSATIONSVERNETZENDE POLYSILOXANMASSEN, EIN HERSTELLUNGSVERFAHREN UND OBERFLÄCHENMODIFIZIERTE FÜLLSTOFFE

(57) Abstract

Condensation cross-linking polysiloxane masses contain at least one cross-linkable polysiloxane, at least one basic filler, at least one phosphorus compound, at least one alkoxysilane cross-linking agent, at least one organometallic compound and if required other auxiliary substances. Also disclosed is a process for preparing these polysiloxane masses, their use, surface-modified fillers and their use.

(57) Zusammenfassung

Die vorliegende Erfindung betrifft kondensationsvernetzende Polysiloxanmassen, enthaltend mindestens ein vernetzungsfähiges Polysiloxan, mindestens einen basischen Füllstoff, mindestens eine Phosphorverbindung, mindestens einen Alkoxysilanvernetzer, mindestens eine metallorganische Verbindung und gegebenenfalls weitere Hilfsstoffe, ein Verfahren zu deren Herstellung sowie deren Verwendung, oberflächenmodifizierte Füllstoffe und deren Verwendung.

Polysiloxane compositions which cross-link by condensation, a process for their production and their use

The present invention relates to polysiloxane compositions which cross-link by condensation and contain at least one cross-linkable polysiloxane, at least one basic filler, at least one phosphorus compound, at least one alkoxysilane cross-linking agent, at least one organometallic compound and optionally other auxiliary substances, and a process for their production and their use.

The polysiloxane compositions according to the invention, referred to hereinafter as RTV-1 (Room-temperature vulcanizing 1-component) alkoxy systems, are compositions which are storable with the exclusion of moisture and cure to form elastomers on exposure to atmospheric moisture with the elimination of alcohols.

Products of this kind were described a long time ago and have become established on the market as sealants. The production of such compositions from OH- or alkoxy-terminated polysiloxanes, optionally unreactive polysiloxane plasticizers, alkoxysilane cross-linking agents, catalysts, fillers and optionally other auxiliary substances is known from US-A 3 294 739, US-A 3 161 614 and US-A 3 494 951.

The material costs of RTV-1 compositions can be considerably reduced by the use of fillers. the fillers do however not only render the compositions less costly but they also substantially influence the properites of the RTV-1 alkoxy systems. The concentration and composition of the fillers used has a crucial effect on the rheological properties of the unvulcanized compositions, such as their stability and flow behaviour. They are furthermore important for the mechanical properties of the cured vulcanizates, such as their tear resistance, elongation at break and modulus of elasticity. Chalks are for example very frequently used as fillers for RTV-1 compositions. Ground natural chalk is usually used together with pyrogenic silica. The silica necessary in this combination if stable compositions are required. Precipitated chalks may be used as an alternative. These chalks can likewise be combined with silica and also with natural chalk. However, in sufficient concentrations, they yield sufficiently stable compositions even without silica. These compositions are distinguished by high tear resistance and elongation

at break as well as good adhesion and they are therefore very suitable for the typical applications in the RTV-1 field, such as for example as sealants.

The use of fillers in RTV-1 compositions, in particular at elevated concentrations, can lead to a high increase in viscosity. This high viscosity can cause difficulties in the production of the compositions and also impair their processing. In addition, at high degrees of filling, the modulus of elasticity of the cured sealants increases and their elongation at break is reduced. Precisely the use of precipitated chalks can lead to very high elastic moduli, which are undesirable for the use of the RTV-1 alkoxy systems as sealants. One further disadvantage which can result from the use of the fillers, such as for example the chalks and in particular the precipitated chalks in RTV-1 alkoxy systems, is the reduced storage life of the unvulcanized pastes. If RTV-1 products are stored with the exclusion of atmospheric moisture and samples are taken from time to time, their cross-linkability can decrease as a function of the storage time. The compositions can even change to such an extent that they completely fail to cross-link upon exposure to atmospheric moisture. RTV-1 alkoxy systems generally have shorter storage lives than other RTV-1 compositions, as a result of which they only have limited applicability.

Some additives are known for various cross-linking systems for improving the rheological properties of highly-filled RTV-1 polysiloxane compositions. Apart from affecting the rheological properties and changing the incorporability of fillers, they also change the mechanical properties of the vulcanizates. They usually reduce the modulus of elasticity. By the addition of such auxiliary substances other important properties of the products can however be affected. Thus, the storage life of the unvulcanized compositions can be shortened and their adhesive properties can be impaired.

Sulphonic acids, and in particular dodecylbenzenesulphonic acid and its salts, are known from EP-A-314 313 and EP-A 314 314 as suitable additives. These sulphonic-acid-containing RTV-1 alkoxy systems result in low moduli of elasticity of the vulcanizates, although the storage life of the unvulcanized products which, without the addition of the sulphonic acid, is relatively short, is not improved by the addition of dodecylbenzenesulphonic acid.

In DE-A-2007002 etherified or esterified polyglycols are used in filled polysiloxane compositions which cross-link by condensation. These products are prepared with the use of various silane cross-linking agents containing at least one nitrogen atom in the molecule, such as for example oximo-, amino- or aminoxysilane cross-linking agents. In DE-A-2 653 499, phosphoric acid esters are described, in combination with the same cross-linking agents. Suitable additives for alkoxy systems that have a low modulus of elasticity and a good storage life are not known.

The problem therefore existed of developing filled RTV-1 alkoxy systems that possess a low modulus of elasticity, high elongation at break and good storage lives in their unvulcanized state. In addition, the resulting cured material should adhere well to many substrates.

Surprisingly, it has now been found that the known disadvantages of highly-filled polysiloxane compositions that cure at room temperature on exposure to atmospheric moisture and release alcohols as cleavage products can be very largely eliminated if phosphoric acid esters and/or polyphosphoric acid esters are added as additives to the compositions. By means of these additives, both the mechanical vulcanizate properties of the RTV-1 alkoxy systems and their storage life in the unvulcanized state are markedly improved.

The present invention provides polysiloxane compositions which cross-link by condensation and contain

a) at least one cross-linkable polysiloxane that contains as reactive terminal group at least one of the following groups

$-O-SiR^1_2OH$, $-O-SiR^1(OR^2)_2$, $-O-Si(OR^2)_3$,

wherein

R^1 denotes optionally substituted C_1-C_8-alkyl, C_6-C_{14}-aryl or C_2-C_8-alkenyl groups and

R^2 denotes optionally substituted linear or branched C_1-C_8-alkyl or C_2-C_8-alkoxyalkyl groups,

and R^1 and R^2 may be the same or different within the molecule,

b) at least one basic filler and optionally other fillers, all of which maybe surface-modified

c) at least one phosphorus compound from the group comprising orthophosphoric acid esters of the following formula I

$$O=P(OR^3)_{3-n}(OH)_n$$

in which

n= 0, 1 or 2 and

R^3= an optionally substituted linear or branched C_1-C_{30}-alkyl, -acyl, C_2-C_{30}-alkenyl or -alkoxyalkyl, C_5-C_{14}-cycloalkyl or C_6-C_{10}-aryl group or a triorganosilyl or diorganoalkoxysilyl group which can be the same or different within the molecule,

and/or esters of polyphosphoric acid,

d) at least one alkoxysilane cross-linking agent of the formula

$$R^1_xSi(OR^2)_{4-x},$$

wherein

x= 0 and 1, and R^1 and R^2 can be the same or different within the molecule,

e) at least one organometallic compound from the group which includes organic dialkyltin(IV) compounds and organic titanates and

f) optionally other auxiliary substances, such as for example plasticizers, bonding agents, stabilizers, pigments, fungicides etc.

Cross-linkable polysiloxanes a) for the purposes of the invention are polydiorganosiloxanes, preferably polydimethylsiloxanes, wherein the methyl groups may optionally be partially replaced by vinyl, phenyl, C_2 to C_8 alkyl or haloalkyl

groups. The polydimethylsiloxanes are substantially linear, but can contain small proportions of organosiloxy units having a branching effect. In a preferred specific embodiment of the present invention the cross-linkable polysiloxane a) has a viscosity between 0.1 and 1000 Pa.s, preferably between 10 and 500 Pa.s. In addition the cross-linkable polydiorganosiloxane a) can be partially substituted by unreactive groups, such as for example trimethylsiloxy groups.

In a preferred specific embodiment of the present invention the reactive groups of the polysiloxanes are

$-O-Si(R^1)_2OH$ groups, in which

R^1 = an optionally substituted C_1-C_8-alkyl, C_6-C_{14}-aryl or C_2-C_8-alkenyl group, and wherein R^1 may be the same or different within the molecule.

The term substituted includes all the usual substituents, such as for example halogen, NO_2, amine, alkoxy, etc.

The basic fillers b) are for example precipitated or ground chalk, metal oxides, sulphates, silicates, hydroxides, carbonates and hydrogencarbonates. Other fillers are e.g. reinforcing and non-reinforcing fillers, such as for example pyrogenic or precipitated silica, carbon black or quartz powder. Both the basic fillers and the other reinforcing or non-reinforcing fillers may optionally be surface-modified. Particularly preferred basic fillers b) are precipitated or ground chalks. Component b) can also consist of mixtures of fillers.

The phosphorus compounds c) according to the invention are esters of ortho- and polyphosphoric acid or mixtures thereof. The esters of orthophosphoric acid are described by the following general formula:

$$O=P(OR^3)_{3-n}(OH)_n,$$

wherein

n = 0, 1 or 2 and

R^3 denotes an optionally substituted linear or branched C_1-C_{30}-alkyl, -acyl, C_2-C_{30}-alkenyl or -alkoxyalkyl, C_5-C_{14}-cycloalkyl or C_6-C_{10}-aryl group or a triorganosilyl or diorganoalkoxysilyl group, and R^3 may be the same or different within the molecule.

In a preferred specific embodiment of the present invention the phosphorus compound c) is an ester of orthophosphoric acid of the formula I, in which R^3 is at least one optionally substituted linear or branched C_4-C_{30}-alkyl group. Where n = 0, R^3 is at least one triorganosilyl or diorganosilyl radical.

Examples of phosphoric acid esters c) according to the invention are primary and secondary esters of orthophosphoric acid as well as mixtures thereof, such as di(2-ethylhexyl) phosphate, dihexadecyl phosphate, diisononyl phosphate, mono-isodecyl phosphate, mono(2-ethylhexyl) phosphate and tris(trimethylsilyl) phosphate.

Component c) can likewise be an ester of polyphosphoric acid or a mixture of several polyphosphoric acid esters. Salts of ortho- and polyphosphoric acid partial esters, such as for example alkali metal salts, also are suitable.

Silane cross-linking agents d) in the polysiloxane compositions according to the invention are alkoxysilanes of the general formula

$$R^1_xSi(OR^2)_{4-x}$$

in which

x = 0 and 1,

R^1 denotes optionally substituted C_1-C_8 alkyl, C_6-C_{14} aryl or C_2-C_8 alkenyl groups and

R^2 denotes optionally substituted C_1-C_8 alkyl, linear or branched C_1-C_8 alkyl or C_2-C_8 alkoxyalkyl groups, wherein R^1 and R^2 can be the same or different within the molecule,

or their mixtures.

Preferred alkoxysilanes are tetraethoxysilane, tetra-n-propoxysilane, methyltriethoxysilane, methyltrimethoxysilane, methyltri(2-methoxyethoxy)silane, vinyltrimethoxysilane or vinyltriethoxysilane. Methyl- and vinyltrimethoxysilane are preferred. Component d) can also be a partial hydrolyzate of the alkoxysilane cross-linking agents.

All catalysts which are commonly used according to the prior art in polysiloxane compositions which cross-link by condensation are suitable as organometallic compounds e). Organic titanium and tin compounds are particularly preferred. Cross-linkable polysiloxanes a) containing the reactive groups

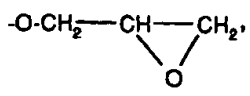
$$-O\text{-}SiR^1(OR^2)_2, \qquad -O\text{-}Si(OR^2)_3$$

are preferably used in combination with organic tin compounds, the groups R^1 and R^2 having the meaning already mentioned. Particularly preferred tin compounds are e.g. diorganotin dicarboxylates, such as dibutyltin dilaurate and dioctyltin maleate as well as solutions of diorganotin oxides in silicic acid esters. Preferred titanium compounds are for example alkyl titanates, such as tetraisopropyl titanate, tetrabutyl titanate and chelated titanium compounds, such as diisobutyl bis(ethyl acetoacetate) titanate, diisopropyl bis(acetylacetonate) titanate or diisopropyl bis(ethylacetoacetate) titanate.

Additives and auxiliary substances f) for the purposes of the invention are preferably plasticizers, bonding agents, pigments and fungicides.

In a preferred specific embodiment of the present invention, the auxiliary substances f) are silicone plasticizers, such as for example polydimethylsiloxanes having terminal trimethylsiloxy groups and a viscosity of 0.1 to 5 Pa.s, bonding agents, such as for example organofunctional silanes of the formulae:

$$X\text{-}CH_2\text{-}CH_2\text{-}CH_2\text{-}Si(OR^2)_3$$

in which X = $-NH\text{-}CH_2\text{-}CH_2\text{-}NH_2$,  ,

$-O\text{-}C(O)\text{-}C(CH_3)=CH_2$, -SH, -OH, -Cl, or NH_2 and

R^2 has the meaning already mentioned above.

The silicone compositions according to the invention preferably consist of

100 parts by weight of a),
10 to 250 parts by weight of b),
0.1 to 25 parts by weight of c),
1 to 30 parts by weight of d)
0.1 to 20 parts by weight of e) and
0 to 240 parts by weight of f).

The total quantity of auxiliary substances and additives f) preferably has the following composition:

0-100 parts by weight of plasticizers,
0-20 parts by weight of bonding agents,
0-100 parts by weight of pigments and
0-20 parts by weight of fungicides,

the sum of all the components f) in the mixture being at most 240 parts by weight.

The present invention also provides a process for the production of the polysiloxane compositions according to the invention, according to which the components a) to f) are mixed with the exclusion of moisture. Preferably the components a), b) and c) are initially introduced and the other components are then added.

The phosphorus compounds c) according to the invention are preferably incorporated into the compositions in the course of the production of the

polysiloxane compositions. In a particularly preferred specific embodiment of the present invention the basic fillers b) and the phosphorus compound c), optionally dissolved in a suitable solvent, are mixed in a preliminary operation. Water or polar or non-polar organic solvents, such as for example alcohols and aromatic or aliphatic hydrocarbons, can for example be used as suitable solvents.

The surface-modified fillers used in the polysiloxane compositions of the present invention are obtained by reacting at least one basic filler b) with at least one phosphorus compound c) from the group comprising orthophosphoric acid esters of the following formula I

$$O = P(OR^3)_{3-n}(OH)_n,$$

in which

n = 0, 1 or 2 and

R^3 = an optionally substituted linear or branched C_1-C_{30}-alkyl, -acyl, C_2-C_{30}-alkenyl or -alkoxyalkyl, C_5-C_{14}-cycloalkyl or C_6-C_{10}-aryl group or a triorganosilyl or diorganoalkoxysilyl group which can be the same or different within the molecule, and/or esters of polyphosphoric acid,

if appropriate in a solvent.

R^3 is preferably C_1-C_{30}-alkyl.

Suitable solvents are water or polar or non-polar organic solvents, such as for example alcohols, aromatic or aliphatic hydrocarbons and/or polydimethylsiloxanes. These surface-modified fillers are hydrophobic.

The present invention also relates to the use of the polysiloxane compositions according to the invention as sealants, adhesives or coating materials.

The present invention also relates to the use of the surface-modified fillers in polysiloxane compositions, plastics, such as for example PVC, thermoplastics, rubber, polysulphide sealants, polyurethane compositions, paints or lacquers.

The following examples serve to illustrate the invention without, however, having any limiting effect.

Examples

General procedure for the preparation and evaluation of the compositions

The compositions were prepared in a 1-litre planetary mixer in accordance with the examples listed hereinafter. After the preparation of the compositions was complete, they were filled into plastic cartridges. The material for the appropriate further tests was taken from the closed cartridges.

The cross-linking behaviour of the polysiloxane compositions was tested on a glass plate, for which purpose the pastes were applied in a layer thickness of 2 mm to an area of 40 x 60 mm. After 24 hours the curing of the material through to the glass surface was examined.

In order to determine the mechanical properties of the vulcanizates, layers of a thickness of 2 mm were produced from the pastes and tested according to DIN 53 504 after being cured for 14 days at 23 °C and 50 % relative atmospheric humidity. The hardness was determined after 21 days according to DIN 53 505.

The storage life of the products was evaluated by storing the pastes in a closed aluminium tube at 50 °C. Samples were taken at one-weekly intervals and tested for cross-linking. If the samples were perfectly cross-linked 1 week after their removal, the test was considered to have been passed. The test for storage life at 50 °C is a method which is commonly used for the evaluation of sealants and which makes it possible to estimate in relatively short periods of time the storage life of the products in practice.

Examples 1 to 8

In a planetary mixer, 44.0 parts by weight of a polydimethylsiloxane containing terminal $Si(CH_3)_2OH$ groups which had a viscosity of 50 Pa.s at 25 °C were mixed with 45.0 parts by weight of a precipitated chalk (BET specific surface area 19 m^2/g) which had been treated with stearic acid, with the addition of various phosphoric acid esters to form a homogeneous paste. The following compounds were used:

Example 1: di-2-ethylhexyl phosphate

Example 2: mono-2-ethylhexyl phosphate

Example 3: monoisodecyl phosphate

Example 4: 1:1 mixture of mono- and di-isononyl phosphate

Example 5: trimethylsilyl-di-2-ethylhexyl phosphate

Example 6: trimethylsilyl-di-2-ethylhexyl phosphate

Example 7: bis-(trimethylsilyl)-mono-2-ethylhexyl phosphate

Example 8: tris-(trimethylsilyl) phosphate

Then 8.0 parts by weight of a polydimethylsiloxane containing terminal $-O-Si(CH_3)_3$ groups and 2.5 parts by weight of methyltrimethoxysilane were stirred in and the composition of the paste was completed by adding 1.0 part by weight of diisobutylbis(ethyl acetoacetate) titanate as well as 0.1 part by weight of N-aminoethyl-3-aminopropyltrimethoxysilane.

The compositions of Examples 1 to 8 cured completely through to the glass plate within 24 hours. The other properties of the compositions are shown in Table 1.

Comparative Example 9

The procedure of Examples 1 to 8 was repeated, the addition of the phosphoric acid ester being omitted. This composition was also completely cured after 24 hours. The testing of the storage life and the mechanical properties did however show that the product only had inadequate properties. The storage life of only 2 weeks at 50 °C and the poor mechanical properties such as high hardness, low elongation and a high modulus of elasticity, very greatly restrict the possible use of such a product, due to the technological disadvantages.

Comparative Example 10

The procedure of Example 9 was repeated, with the addition of 1.5 parts by weight of dodecylbenzenesulphonic acid. The sealant had cured through

completely after 24 hours. The storage life of the product does not differ from that of Comparative Example 5 without any additive and, since it is only 2 weeks at 50 °C, is inadequate. The mechanical properties of the vulcanizate are of a high standard and include high elongation at break and a low modulus of elasticity (Table 1).

Table 1: Test results of Examples 1 to 10

Example No.	Parts by weight of phosphate	Shore A hardness	Elongation at break [%]	Modulus of elasticity [N/mm²]	Tear resistance [N/mm²]	Storage life[1] at 50 °C [in weeks]
1	1.5	28	880	0.49	2.0	4
2	1.0	17	820	0.41	1.7	10
3	1.1	23	780	0.45	1.7	10
4	1.3	23	780	0.46	1.8	10
5	1.0	33	630	0.63	2.0	6
6	1.25	31	930	0.50	2.34	6
7	1.0	28	830	0.50	2.11	14
8	1.0	30	500	0.45	1.99	14
9[2]	-	35	640	0.67	2.6	2[3]
10[2]	1.5[4]	24	930	0.44	2.0	2[3]

1) after the indicated time the products were still in a perfect condition

2) comparative example

3) the composition no longer cures after 3 weeks. The product is highly damaged.

4) dodecylbenzenesulphonic acid.

THE CLAIMS DEFINING THE INVENTION ARE AS FOLLOWS:

1. Polysiloxane compositions which cross-link by condensation and contain

a) at least one cross-linkable polysiloxane, that contains as a reactive terminal group at least one of the following groups

$-O-SiR^1_2OH$, $-O-SiR^1(OR^2)_2$, $-O-Si(OR^2)_3$,

wherein

R^1 denotes optionally substituted C_1-C_8-alkyl, C_6-C_{14}-aryl or C_2-C_8-alkenyl groups and

R^2 denotes optionally substituted linear or branched C_1-C_8-alkyl or C_2-C_8-alkoxyalkyl groups, and R^1 and R^2 can be the same or different within the molecule,

b) at least one basic filler and optionally other fillers, all of which maybe surface-modified,

c) at least one phosphorus compound from the group comprising orthophosphoric acid esters of the following formula I

$O=P(OR^3)_{3-n}(OH)_n$,

in which

n = 0, 1 or 2 and

R^3 = an optionally substituted linear or branched C_1-C_{30}-alkyl, -acyl, C_2-C_{30}-alkenyl or -alkoxyalkyl, C_5-C_{14}-cycloalkyl or C_6-C_{12}-aryl group or a triorganosilyl or diorganoalkoxysilyl group which can be the same or different within the molecule, and/or an ester of polyphosphoric acid,

d) at least one alkoxysilane cross-linking agent of the formula

$R^1_xSi(OR^2)_{4-x}$,

wherein

x = 0 and 1, and

R^1 and R^2 can be the same or different within the molecule,

e) at least one organometallic compound from the group which includes organic dialkyltin(IV)- compounds and organic titanates, and

f) optionally other auxiliary substances.

2. Polysiloxane compositions which cross-link by condensation, according to Claim 1, characterized in that the cross-linkable polysiloxane a) has a viscosity of between 0.1 and 1000 Pa.s.

3. Polysiloxane compositions which cross-link by condensation, according to one of Claims 1 and 2, characterized in that the basic fillers b) are precipitated or ground chalks.

4. Polysiloxane compositions which cross-link by condensation, according to anyone of Claims 1 to 3, characterized in that the phosphorus compound c) is

an ester of orthophosphoric acid containing at least one optionally substituted linear or branched C_4-C_{30}-alkyl group R^3.

5. Polysiloxane compositions which cross-link by condensation, according to anyone of Claims 1 to 4, characterized in that the alkoxysilane cross-linking agent d) is tetraethoxysilane, tetra-n-propoxysilane, methyl-triethoxysilane, methyltrimethoxysilane, methyltri(2-methoxyethoxy)-silane, vinyltrimethoxysilane or vinyltriethoxysilane.

6. Polysiloxane compositions which cross-link by condensation, according to anyone of Claims 1 to 5, characterized in that the organometallic compound e) is an organic titanium or tin compound.

7. Polysiloxane compositions which cross-link by condensation, according to anyone of Claims 1 to 6, characterized in that the auxiliary substances f) are plasticizers, catalysts, bonding agents, pigments and/or fungicides.

8. Polysiloxane compositions which cross-link by condensation, according to anyone of Claims 1 to 7, characterized in that they consist of

100 parts by weight of a),
10 to 250 parts by weight of b),
0.1 to 25 parts by weight of c),
1 to 30 parts by weight of d),
0.1 to 20 parts by weight of e) and
0 to 240 parts by weight of f).

9. Polysiloxane compositions which cross-link by condensation, according to anyone of Claims 1 to 7, characterized in that the auxiliary substance f) has the following composition:

0-100 parts by weight of plasticizers,
0-20 parts by weight of bonding agents,
0-100 parts by weight of pigments,
0-20 parts by weight of fungicides,

the sum of all the components f) in the mixture amounting to a maximum of 240 parts by weight.

10. Process for the production of the polysiloxane compositions which cross-link by condensation, according to anyone of Claims 1 to 9, wherein the basic fillers b) and the phosphorus compound c), optionally dissolved in a suitable solvent, are mixed in a preliminary operation, prior to mixing with components a), d), e) and f) in the absence of moisture.

11. Polysiloxane compositions which cross-link by condensation according to anyone of Claims 1 to 9 wherein said surface-modified fillers are obtained by reacting at least one basic filler b) with at least one phosphorus compound c) from the group comprising orthophosphoric acid esters of the following formula I

$$O=P(OR^3)_{3-n}(OH)_n,$$

in which

n = 0, 1 or 2 and

R^3 an optionally substituted linear or branched C_1-C_{30}-alkyl, -acyl, C_2-C_{30}-alkenyl or -alkoxyalkyl,
C_5-C_{14}-cycloalkyl or C_6-C_{10}-aryl group or a triorganosilyl or diorganoalkoxysilyl group which can be the same or different within the molecule, and/or esters of polyphosphoric acid,

if appropriate in a solvent.

12. Use of the surface-modified fillers set forth in claim 11 in polysiloxane compositions, plastics, paints or lacquers.

13. Use of the polysiloxane compositions which cross-link by condensation, according to anyone of Claims 1 to 9, as sealants, adhesives or coating compositions.

DATED this 22nd day of April, 1998.

BAYER AG

By Its Patent Attorneys

DAVIES COLLISON CAVE